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*IPe*  
PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of:

ANATOLY CHEKHMIR, ET AL.

Serial No.: 10/606,218

ATTN. APPLICATION BRANCH

Filed: June 26, 2003

For: PROCESS FOR IMMOBILIZING  
RADIOACTIVE AND  
HAZARDOUS WASTES

**CLAIM TO PRIORITY UNDER 35 U.S.C. § 119**

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

The benefit of the filing date of the following prior application filed in the following foreign country is hereby requested and the right of the priority provided under 35 U.S.C. § 119 is hereby claimed:

Israeli Patent Appln. No. 136,685 filed June 12, 2000

In support of this claim, filed herewith is a certified copy of said foreign application.

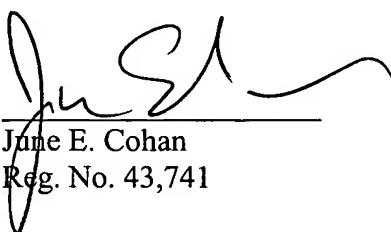
Respectfully submitted

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This 16-08-2005 היום

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חוק הפטנטים, התשכ"ז - 1967  
PATENTS LAW, 5727-1967

## בקשה לפטנט

Application for Patent

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בעל האמצאה מכח העברה ששם הוא:  
Owner of an invention, by virtue of an assignment, the title of which is:

### תהליך לטיפול בפסולת גרעינית

(בעברית)  
(Hebrew)  
(באנגלית)  
(English)

### PROCESS FOR TREATMENT OF RADIOACTIVE WASTE

hereby apply for a patent to be granted to me in respect

מבקש בזאת כי יינתן לי עליה פטנט.  
thereof.

* בקשה חילוקה - Application for Division		* בקשה פטנט מוסף - Application for Patent of Addition		* דרישת דין קדימה Priority Claim		
מבקש פטנט from Application	No. _____ מס' _____ dated _____ יום _____	לבקשת פטנט to Patent/Appl.	No. _____ מס' _____ dated _____ יום _____	מספר/סימן Number/Mark	תאריך Date	מדינת האיחוד Convention Country
<b>* ייפוי כללי מוגש בזאת</b> General P.O.A: enclosed herewith						
המע למסירתה הוודאות ומסמכים בישראל Address for Service in Israel ייחודה מאיר ת.ד. 130 71700 מודיעין						
חתימת המבקש Signature of Applicant						
היום 11 בחודש יוני שנת 2000 This 11 th of June 2000						

לשימוש הלשכה  
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טופס זה, כשהוא מוטבע בחותם לשכת הפטנטים ומוסלם במשטר ובתאריך ההגשתה, הינו אישור להגשת הבקשה שפרטיה רשומים לעיל.

This form, impressed with the Seal of the Patent Office and indicating the number and date of filing, certifies the filing of the application, the particulars of which are set out above

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תהליך לטיפול בפסולת גרעינית

**PROCESS FOR TREATMENT OF RADIOACTIVE WASTE**

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## PROCESS FOR TREATMENT OF RADIOACTIVE WASTE

### FIELD OF THE INVENTION

The field of the invention is immobilization of radioactive wastes.

### BACKGROUND OF THE INVENTION

The use of radioactive compounds in the industry leads to the acute need for useful means of disposal of the waste of said radioactive compounds. Thus, there is a need for an efficient, comprehensive, process for immobilizing radioactive waste.

### SUMMARY OF THE INVENTION

According to a first aspect, referred to hereinafter as "aspect I" the present invention relates to a process for immobilizing radioactive wastes comprising the steps of:

- a) bonding a radionuclide to a host mineral;
- b) covering the particles resulting from step (a) with the same host mineral by epitaxy; and
- c) covering the result of step (b) with a rock; said rock comprising components of said host mineral.

According to a second aspect, referred to hereinafter as "aspect II", the present invention relates to a process for immobilizing radioactive wastes comprising the steps of:

- a) bonding a radionuclide to a host mineral;

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- b) sintering the result of the previous step with additional host mineral, this step being performed once or twice, as will be explained hereinafter; and
- c) covering the result of step (b) with a rock, said rock comprising components of said host mineral.

#### **DETAILED DESCRIPTION OF EMBODIMENTS OF THE INVENTION**

The term "host minerals" is defined throughout this document as minerals in which radionuclides may be incorporated.

The term "flux" is defined throughout this document as any chemical substance used to reduce the temperature of a process.

The term "nuclide" is defined throughout this document as a chemical substance in which essentially all atoms have the same atomic number and mass number.

The term "radionuclide" is defined throughout this document as a radioactive nuclide.

The term "activation" when referring to host minerals is defined throughout this document as any process which enhances the capability of host minerals to absorb radionuclides.

The term "active" when referring to host minerals is defined throughout this document as capable of absorbing radionuclides.

The term "epitaxy" is defined throughout this document as an oriented overgrowth of crystalline material upon the surface of a host mineral of slightly different chemical composition but similar structure.

The term "sintering" is defined throughout this document as the thermal treatment of a powder or compact at a temperature below the melting point of the main constituent, for the purpose of increasing its strength by bonding together of the particles.

The term "effective covering" in the context of covering host mineral containing radionuclides is defined throughout this document as covering host mineral containing radionuclides so that there is no chemical interaction between the host mineral containing radionuclides and the biosphere.

*[absorption]* According to one embodiment the bonding of the radionuclide to the host mineral in step (a) is achieved by bringing a solution of a radionuclide in a solvent in contact with an active host mineral. This can be performed by stirring the radionuclide with the host mineral, by filtering the radionuclide solution through the host mineral or by any other absorption method known in the art.

*[synthesis 1]* According to another embodiment the bonding of the radionuclide to the host mineral in step (a) is achieved by mixing oxides of the components of the host mineral with oxides of the radionuclides at a temperature of up to 1200°C.

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[synthesis 2] According to another embodiment the bonding of the radionuclide to the host mineral in step (a) is achieved by mixing oxides of the components of the host mineral with radionuclides dissolved in a solvent at a temperature up to 1200°C.

In all these 3 embodiments, the final product is a host mineral in which the radionuclide-oxides have replaced cations of the host mineral.

[epitaxy] According to one embodiment, the covering of step (b) in aspect I may be achieved by the steps of:

- 1) mixing the result of step (a) of aspect I with rock or flux and heating to a temperature of up to 1000°C, said rock or flux comprising components of said host mineral;
- 2) allowing for an effective covering of the host mineral to form on said result of step (a).

According to this embodiment, step 2) may be achieved by cooling the rock or flux at a constant or inconstant rate or essentially maintaining the temperature achieved in step (1).

According to this embodiment if flux was used, the flux is then removed at this point by dissolving the result of step 2) either in boiling water or in another solvent and then separating the host mineral from the boiling water or the solvent which now contains the components of the flux.

In the event that flux was used, the result is encapsulated crystals that must be put in a matrix. This is done by mixing the crystals with melted rock that is the same as that found in the disposal site and which contains the host mineral. This mixture is then heated above the melting point of the rock but below the melting point of the crystals (between 800°C and 1200°C).

The product of step (b) of aspect II subsequent to the performance of said step (b) is a high-density ceramic in which crystals containing radionuclides are surrounded by the same crystals without radionuclides. However, subsequent to the performance of said step (b) the first time, there still remain some radioactive crystals on the surface of said ceramic.

The product of step (b) of aspect II subsequent to the second performance of said step (b) is a high-density ceramic in which there are essentially no crystals containing radonuclides on the surface.

According to an embodiment of the invention, the result of steps (b) and (c) can be obtained by performing one step if the rock matrix has sufficient amount of the components of the host mineral to produce a thick epitaxial layer.

*[disposal site rock covering]* According to another embodiment, the covering of the result of step (b) may be achieved by the steps of:

- 1) mixing the result of step (b) with rock at temperature up to 1200°C, said rock containing components of said host mineral; and

- 2) allowing for an effective covering of the rock to form on the host mineral in such a way that no radionuclides are in contact with the surrounding environment.

According to this embodiment, step 2) may be achieved by cooling the rock at a constant or inconstant rate or essentially maintaining the temperature achieved in step (1).

According to this embodiment the result is encapsulated crystals containing radionuclides in a rock matrix.

The host minerals used in the present invention are typically but not limitatively selected from the group that consists of phosphates, silicates and clays.

The rocks used in the present invention are typically but not limitatively selected from the group that consists of granitoid, dacite, andesite, basalt, ultrabasic rocks, carbonatites, amphibolites and clays.

Some host minerals cannot retain radionuclides in natural conditions in which case contact with solvents will cause the radionuclides to be removed from the host minerals. These host minerals must be transformed to minerals that can retain radionuclides. In such cases, this transformation is performed subsequent to step (a) by heating the host mineral to a temperature up to 1000°C. Thus zeolites must be transformed into feldspar by heating at a temperature between 600°C and 1000°C for up to 1 day.

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Preferably, the result of step (a) is washed in water and dried in heat prior the performance of step (b), preferably at about 400°C.

In all instances that rock is used for covering, it is advisable to use a type of rock similar or identical to the type of rock present in the burial site in order to add to the stability of the rock layer of the present invention.

### EXAMPLES

The following examples are intended to illustrate certain embodiments of the present invention, and by any means should not be used in order to limit the scope of the claims.

#### Example 1

A synthetic Na-zeolite was crushed and heated to about 400°C for about 12 hours. Cerium was dissolved in nitric acid resulting in a liquid containing Cerium. This liquid was then filtered through the Na-zeolite at room temperature. The Na-zeolite was then washed in water at room temperature and then dried at 400°C for about 12 hours. It was then heated for about 6 hours at about 800°C resulting in the Na-zeolite being transformed into feldspar containing Cerium. It was then mixed with sodium tetraborite containing feldspar. The result was heated until melted for about 36 hours at about 950°C-1000°C and cooled by gradual decrease of temperature to 400°C for 8 hours. The sodium tetraborite was then removed with boiling water, leaving crystals with Cerium inside covered with a feldspar layer. Crushed obsidian was mixed with

the covered crystals and heated at about 1000°C for several hours and then cooled by gradual decrease in temperature to 400°C for 8 hours, resulting in an obsidian matrix which contained feldspar crystals containing Cerium covered by a feldspar layer.

### Example 2

$\text{Ce}_2\text{O}_3$  and  $\text{NaPO}_3$  and  $\text{La}_2\text{O}_3$  (the components of monazite) were taken and about 10-15% of the Cerium was replaced by Uranium and Thorium. This was done when all of these components were heated together at 1200°C for about 2 days. The result was synthesized monazite in which part of the Cerium and Lantanium were replaced by Uranium and Thorium. The synthesized monazite was then quenched. It was then mixed with  $\text{Na}_2\text{B}_4\text{O}_7 \cdot 10\text{H}_2\text{O}$  containing  $\text{Ce}_2\text{O}_3 + \text{P}_2\text{O}_5$ . This was then heated at 900°C for about 2 days. This was then cooled to room temperature for 8 hours by turning off the furnace. Next the  $\text{Na}_2\text{B}_4\text{O}_7 \cdot 10\text{H}_2\text{O}$  was dissolved in boiling water, leaving monazite crystals with Uranium and Thorium inside covered with monazite without Uranium and Thorium. These crystals were then mixed with crushed obsidian and melted at 1000°C for several hours, and then cooled by gradual decrease of temperature to 400°C for 8 hours. This resulted in a block of obsidian with the encapsulated monazite crystals inside.

### Example 3

$\text{Ce}_2\text{O}_3$  and  $\text{NaPO}_3$  and  $\text{La}_2\text{O}_3$  (the components of monazite) were taken and about 10-15% of the Cerium was replaced by Uranium and Thorium. This was done when all of these components were heated together at 1200°C for about 2 days. The result was synthesized monazite in which part of the Cerium and Lantanium were replaced by Uranium and Thorium. The synthesized monazite was then quenched. The

synthesized monazite containing the Uranium and Thorium was mixed with natural monazite of the same composition not containing Uranium and Thorium. About 25% of the synthesized monazite was mixed with 75% of the natural monazite. This mixture was heated to about 900°C-1000°C for about 1 day and then was cooled by gradual decrease of temperature to room temperature for 8 hours. At this point crystals were obtained containing Uranium and Thorium surrounded by crystals without Uranium and Thorium. Some Uranium and Thorium crystals remained on the surface. The crystals were added to crushed monazite and heated to about 900°C-1000°C for one half hour resulting in the addition of a monazite layer covering the synthesized monazite. This was then cooled by gradual decrease of temperature to room temperature for 8 hours. The result was mixed them crushed obsidian and melted at about 1000°C for about 1 day. This was then cooled by gradual decrease of temperature to room temperature for 8 hours.

**CLAIMS**

- 1) A process for immobilizing radioactive wastes comprising the steps of:
  - a) bonding a radionuclide to a host mineral;
  - b) covering the result of step (a) with the same host mineral, by epitaxy; and
  - c) covering the result of step (b) with a rock, said rock comprising components of said host mineral.
- 2) A process for immobilizing radioactive wastes comprising the steps of:
  - a) bonding a radionuclide to a host mineral;
  - b) sintering the result of the previous step with additional host mineral; and
  - c) covering the result of step (b) with a rock, said rock comprising components of said host mineral.
- 3) A process for immobilizing radioactive wastes comprising the steps of:
  - a) bonding a radionuclide to a host mineral;
  - b) sintering the particles resulting from the previous step with additional host mineral and further sintering the sintered material with additional host mineral; and
  - c) covering the result of step (b) with a rock, said rock comprising components of said host mineral.
- 4) A process according to any of claims 1-3 in which step (a) is achieved by absorbing the radionuclide to the mineral.

- 5) A process according to claim 4 in which the absorption is performed by stirring a solution comprising the radionuclides with the host mineral.
- 6) A process according to claim 4 in which the absorption is performed by filtering a solution containing the radionuclides through the host mineral.
- 7) A process according to any of claims 1-3 in which step (a) is achieved by mixing oxides of the components of the host mineral with oxides of the radionuclides at a temperature of up to 1200°C.

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- 8) A process according to any of claims 1-3 in which step (a) is achieved by mixing oxides of the components of the host mineral with radionuclides dissolved in a solvent at a temperature up to 1200°C.
- 9) A process according to any of claims 1-3 in which the result of step (a) of any of claims 1-3 is heated to a temperature up to 1000°C prior to the performance of the corresponding step (b)
- 10) A process according to claim 1 in which step (b) is achieved by the steps of:
  - (I) mixing the result of step (a) of claim (1) with rock or flux and heating to a temperature of up to 1000°C, said rock or flux comprising components of said host mineral; and
  - (II) allowing for an effective covering of the host mineral to form on said result of step (a).

11) A process according to claim 10 in which step (II) is performed by any method selected from:

- a) cooling the rock or flux at a constant rate;
- b) cooling the rock or flux at an inconstant rate; and
- c) maintaining the temperature achieved in step (I).

12) A process according to any of the preceding claims wherein in the event that flux is used, it is removed by dissolving the flux in boiling water or in another solvent in which the flux is soluble and the host mineral is essentially insoluble and separating the host mineral from the solution comprising flux.

13) A process according to any of the preceding claims wherein in the event that flux is used, the result of step (II) in claim (10) is mixed with rock and heated to a temperature that is above the melting point of the rock but below the melting point of said result of step (II) of claim (10).

14) A process according to claim 13 in which the mixture is heated to a temperature of between 800°C and 1200°C.

15) A process according to any of the preceding claims in which step (c) is performed by the steps of:

- (I) mixing the result of step (b) with rock, and heating to a temperature up to 1200°C, said rock containing said host mineral; and
- (II) allowing for an effective covering of the rock to form on the host mineral.

16) A process according to claim 15 in which step (II) is performed by any method selected from:

- a) cooling the rock or flux at a constant rate;
- b) cooling the rock or flux at an inconstant rate; and
- c) maintaining the temperature achieved in step (I).

17) A process according to any of the preceding claims in which the host minerals are one or more selected from the group that consists of phosphates, silicates and clays.

18) A process according to any of the preceding claims in which the rock is one or more rocks selected from the group that consists of granitoid, dacite, andesite, basalt, ultrabasic rocks, carbonatites, amphibolites and clays.

19) A process according to any of the preceding claims in which steps (b) and (c) of claim 1 are combined to one step by adding an additional amount of the components of the host mineral to produce a thick epitaxial layer.

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